

## QUINTINITE-2H, QUINTINITE-3T, CHARMARITE-2H, CHARMARITE-3T AND CARESITE-3T, A NEW GROUP OF CARBONATE MINERALS RELATED TO THE HYDROTALCITE – MANASSEITE GROUP

GEORGE Y. CHAO\*

*Ottawa–Carleton Geoscience Centre, Department of Earth Sciences, Carleton University, Ottawa, Ontario K1S 5B6*

ROBERT A. GAULT<sup>1</sup>

*Research Division, Canadian Museum of Nature, Ottawa, Ontario K1P 6P4*

### ABSTRACT

Charmarite-2H, charmarite-3T, quintinite-2H, quintinite-3T and caresite-3T are a group of new carbonate minerals with the general formula  $M^{2+}_4Al^{3+}_2(OH)_{12}CO_3 \cdot 3H_2O$  (4:2 minerals). They are structurally and chemically related to the hydrotalcite–manasseite group (6:2 minerals). Superlattice reflections in the single-crystal X-ray photographs suggest ordering of cations in the 4:2 minerals, whereas the cations of the 6:2 minerals are disordered. 2H and 3T designate the 2-layered hexagonal and 3-layered trigonal structures, respectively. All five species are late-stage hydrothermal minerals in alkaline rocks, and are associated with many rare mineral species. Quintinite-2H occurs at the Jacupiranga mine, São Paulo, Brazil; charmarite-2H, charmarite-3T and quintinite-3T occur at Mont Saint-Hilaire, Quebec; caresite-3T occurs at Mont Saint-Hilaire and at the Corporation Quarry, Montreal, Quebec. In habit, they range from thin plates to thick tabular crystals, in some cases forming pyramidal or rosette-like aggregates. They generally range from yellow to brown, occasionally colorless to pale blue. All are transparent, with a white streak and vitreous luster, are non-fluorescent, and have a Mohs hardness of approximately 2. All are brittle, with a perfect {001} cleavage and uneven fracture. The measured densities are: charmarite-2H 2.47(1), charmarite-3T 2.48(1), quintinite-2H and quintinite-3T 2.14(1), caresite-3T 2.59(1) g/cm<sup>3</sup>. Charmarite-2H is uniaxial negative,  $\omega = 1.587(1)$ ,  $\epsilon = 1.547(1)$ ; charmarite-3T is uniaxial negative,  $\omega = 1.587(1)$ ,  $\epsilon$  undetermined; quintinite-2H is uniaxial positive,  $\omega = 1.533(1)$ ,  $\epsilon = 1.533(1)$ ; quintinite-3T is uniaxial positive or negative,  $\omega = 1.533(1)$ ,  $\epsilon = 1.533(1)$ ; caresite-3T is uniaxial negative,  $\omega = 1.599(1)$ ,  $\epsilon = 1.570(1)$ . All are generally pleochroic, with only the near-end-member quintinite-3T being non-pleochroic. Charmarite-2H is hexagonal, space group  $P6_322$ ,  $a$  10.985(5),  $c$  15.10(2) Å,  $V$  1578(3) Å<sup>3</sup>,  $Z = 4$ ; charmarite-3T is trigonal,  $P3_112$  or  $P3_212$ ,  $a$  10.985(3),  $c$  22.63(3) Å,  $V$  2366(4) Å<sup>3</sup>,  $Z = 6$ ; quintinite-2H is hexagonal,  $P6_322$ ,  $a$  10.571(1),  $c$  15.139(7) Å,  $V$  1465(1) Å<sup>3</sup>,  $Z = 4$ ; quintinite-3T is trigonal,  $P3_112$  or  $P3_212$ ,  $a$  10.558(2),  $c$  22.71(3) Å,  $V$  2192(3) Å<sup>3</sup>,  $Z = 6$ ; caresite-3T is trigonal,  $P3_112$  or  $P3_212$ ,  $a$  10.805,  $c$  22.48(3) Å,  $V$  2273(4) Å<sup>3</sup>,  $Z = 6$ . The following ideal formulae were developed from electron-microprobe analysis coupled with TGA–EGA data in some cases: charmarite-2H and charmarite-3T:  $Mn_4Al_2(OH)_{12}CO_3 \cdot 3H_2O$ ; quintinite-2H and quintinite-3T:  $Mg_4Al_2(OH)_{12}CO_3 \cdot 3H_2O$ ; caresite-3T:  $Fe_4Al_2(OH)_{12}CO_3 \cdot 3H_2O$ . IR spectra are given for all five species.

**Keywords:** charmarite-2H, charmarite-3T, quintinite-2H, quintinite-3T, caresite-3T, new mineral species, hydrotalcite, manasseite, Jacupiranga mine, Brazil, Mont Saint-Hilaire, Corporation Quarry, Montreal, Quebec.

### SOMMAIRE

Les nouvelles espèces charmarite-2H, charmarite-3T, quintinite-2H, quintinite-3T et caresite-3T constituent un groupe de minéraux carbonatés ayant la formule chimique générale  $M^{2+}_4Al^{3+}_2(OH)_{12}CO_3 \cdot 3H_2O$  (minéraux 4:2). Ils sont structuralement et chimiquement apparentés au groupe de la hydrotalcite – manasséite group (minéraux 6:2). La présence de réflexions d'une surstructure dans les clichés de précession X indiquent une mise en ordre dans les minéraux 4:2, tandis que les cations sont désordonnés dans le cas des minéraux 6:2. Les symboles 2H et 3T indiquent les structures hexagonale, à deux couches, et trigonale, à trois couches, respectivement. Les cinq espèces sont tardives et d'origine hydrothermale dans des roches alcalines, et sont associées à plusieurs espèces rares. La quintinite-2H a été découverte à la mine Jacupiranga, province de São Paulo, Brésil; la charmarite-2H, la charmarite-3T et la quintinite-3T ont été découvertes au mont Saint-Hilaire, Québec; la caresite-3T est présente aussi bien au mont Saint-Hilaire qu'à la carrière Corporation, à Montréal, Québec. Ces minéraux se présentent en plaquettes minces ou épaisses, et en certains cas en amas pyramidaux ou en rosette. En général, ce sont des minéraux jaunes à bruns, et plus rarement, incolores à bleu pâle. Ils sont transparents, avec une rayure blanche et un éclat vitreux; ils ne sont pas fluorescents, et ont une dureté de Mohs d'environ 2. Tous sont cassants, avec un clivage {001} parfait

\* Present address: 2031 Delmar Drive, Ottawa, Ontario K1H 5P6.

<sup>1</sup> E-mail address: rgault@mus-nature.ca

et une cassure inégale. Les densités mesurées sont: charmarite-2H 2.47(1), charmarite-3T 2.48(1), quintinite-2H et quintinite-3T 2.14(1), caresite-3T 2.59(1) g/cm<sup>3</sup>. La charmarite-2H est uniaxe négative,  $\omega = 1.587(1)$ ,  $\epsilon = 1.547(1)$ ; la charmarite-3T est uniaxe négative,  $\omega = 1.587(1)$ ,  $\epsilon$  non déterminé. La quintinite-2H est uniaxe positive,  $\omega = 1.533(1)$ ,  $\epsilon = 1.533(1)$ ; la quintinite-3T est uniaxe positive ou négative,  $\omega = 1.533(1)$ ,  $\epsilon = 1.533(1)$ . La caresite-3T est uniaxe négative,  $\omega = 1.599(1)$ ,  $\epsilon = 1.570(1)$ . Tous sont pléochroïques, en général; seule la quintinite-3T dont la composition est presque celle du pôle est non pléochroïque. La charmarite-2H est hexagonale, groupe spatial  $P6_322$ ,  $a$  10.985(5),  $c$  15.10(2) Å,  $V$  1578(3) Å<sup>3</sup>,  $Z = 4$ ; la charmarite-3T est trigonale,  $P3_12$  ou  $P3_212$ ,  $a$  10.985(3),  $c$  22.63(3) Å,  $V$  2366(4) Å<sup>3</sup>,  $Z = 6$ . La quintinite-2H est hexagonale,  $P6_322$ ,  $a$  10.571(1),  $c$  15.139(7) Å,  $V$  1465(1) Å<sup>3</sup>,  $Z = 4$ ; la quintinite-3T est trigonale,  $P3_12$  ou  $P3_212$ ,  $a$  10.558(2),  $c$  22.71(3) Å,  $V$  2192(3) Å<sup>3</sup>,  $Z = 6$ . La caresite-3T est trigonale,  $P3_12$  ou  $P3_212$ ,  $a$  10.805,  $c$  22.48(3) Å,  $V$  2273(4) Å<sup>3</sup>,  $Z = 6$ . Les formules idéales suivantes découlent des résultats d'analyses à la microsonde électronique, de données thermogravimétriques et, dans certains cas, d'analyse des gaz évolués: charmarite-2H et charmarite-3T:  $Mn_4Al_2(OH)_{12}CO_3 \cdot 3H_2O$ ; quintinite-2H et quintinite-3T:  $Mg_4Al_2(OH)_{12}CO_3 \cdot 3H_2O$ ; caresite-3T:  $Fe_4Al_2(OH)_{12}CO_3 \cdot 3H_2O$ . Les spectres de transmission dans l'infrarouge sont présentés pour les cinq espèces.

(Traduit par la Rédaction)

**Mots-clés:** charmarite-2H, charmarite-3T, quintinite-2H, quintinite-3T, caresite-3T, nouvelles espèces minérales, hydrotalcite, manasséite, mine de Jacupiranga, Brésil, mont Saint-Hilaire, carrière Corporation, Montréal, Québec.

## INTRODUCTION

A new group of carbonate minerals with the general formula  $M^{2+}_4Al^{3+}_2(OH)_{12}CO_3 \cdot 3H_2O$  (or 4:2 minerals) includes charmarite-2H and charmarite-3T in which  $M = Mn$ , quintinite-2H and quintinite-3T in which  $M = Mg$ , and caresite-3T in which  $M = Fe$ . The 2H form of caresite-3T has not yet been found. This group of minerals was formerly designated as UK 58 (Chao *et al.* 1990) and are chemically and structurally related to the hydrotalcite and manasseite groups:  $M^{2+}_6M^{3+}_2(OH)_{16}CO_3 \cdot 4H_2O$  (or 6:2 minerals) where  $M^{2+} = Mg$  and  $Ni$ , and  $M^{3+} = Cr$ ,  $Fe$ ,  $Al$ ,  $Co$  and  $Mn$ . Charmarite-2H, charmarite-3T, quintinite-3T and caresite-3T occur in the quarries at Mont Saint-Hilaire, Rouville County, Quebec. Quintinite-2H is found in the Jacupiranga mine, São Paulo, Brazil. Caresite-3T also occurs in construction excavations in the old Corporation quarry, Mont Royal, Montreal, Quebec. Charmarite (pronounced CHAR • MĀRAIT) is named for Charles H. (b. 1917) and Marcelle (b. 1918) Weber, amateur mineralogists from Guilford, Connecticut, co-finders of the 2H form of the mineral. Quintinite (KWIN • TĪ • NAIT) is named for Quintin Wight (b. 1935), of Ottawa, Ontario, author of *The Complete Book of Micromounting* (1993). Caresite (KĒR • SAIT) is named for Stephen (b. 1909) and Janet (b. 1921) Cares, amateur mineralogists from Sudbury, Massachusetts, co-finders of the mineral. All are mineral collectors who have contributed significantly to the study of the minerals from Mont Saint-Hilaire. The suffixes 2H and 3T denote the two-layered hexagonal and three-layered trigonal forms, respectively. These new minerals and their names have been approved by the Commission on New Minerals and Mineral Names, IMA. Cotype material is housed in the collection of the Canadian Museum of Nature, Ottawa, under catalogue numbers CMNMI 81542, CMNMI 81543 and CMNMI 81544 (charmarite-2H); CMNMI 81545 (charmarite-3T); CMNMI 81546, CMNMI 47266 and CMNMI 81548

(quintinite-2H); CMNMI 81549 (quintinite-3T); CMNMI 81550, CMNMI 81551 and CMNMI 81552 (caresite-3T), and in the collection of the Royal Ontario Museum, Toronto, under catalogue numbers M45194, M46765 and M46766 (charmarite-2H); M46767 (charmarite-3T); M46768, M46769 and M46770 (quintinite-2H); M46771 (quintinite-3T); M46772, M46773 and M46774 (caresite-3T). In this paper, we present the physical and chemical data that establish these species as new minerals in a group that is distinct from the hydrotalcite–manasseite group.

## OCCURRENCE

Charmarite-2H, charmarite-3T, quintinite-3T and caresite-3T were found in the alkaline complex of Mont Saint-Hilaire, Quebec, one of ten related complexes collectively known as the Monteregian Hills. Mont Saint-Hilaire is well known for its large suite of rare minerals (Horváth & Gault 1990, Mandarino & Anderson 1989), which now numbers 313 known species. The new species described herein occur as late-stage hydrothermal minerals in miarolitic cavities and in pegmatitic bodies within nepheline syenite. Charmarite-2H was collected by the Webers in 1971 and 1976 from the Demix quarry. Other specimens were collected by Jan Van Luling in 1981 from the Demix quarry and by Stephen and Janet Cares in 1988 from the Poudrette quarry. Charmarite-3T was collected by the Cares in 1975 from the Demix quarry. Both polytypes of charmarite are associated with analcime, natrolite, microcline, aegirine, astrophyllite, tetranatrolite, catapleite, calcite, siderite, rhodochrosite, burbankite, kutnohorite and an amorphous phase. Quintinite-3T was collected from the second level of the Demix quarry in 1973 by László and Elsa Horváth. It is associated with analcime, natrolite, tetranatrolite, aegirine, calcite, siderite, fluorite, pyrite, biotite, donnayite-(Y) and ancylite-(Ce). In another occurrence, trace amounts of quintinite-3T were identified in aggregates of

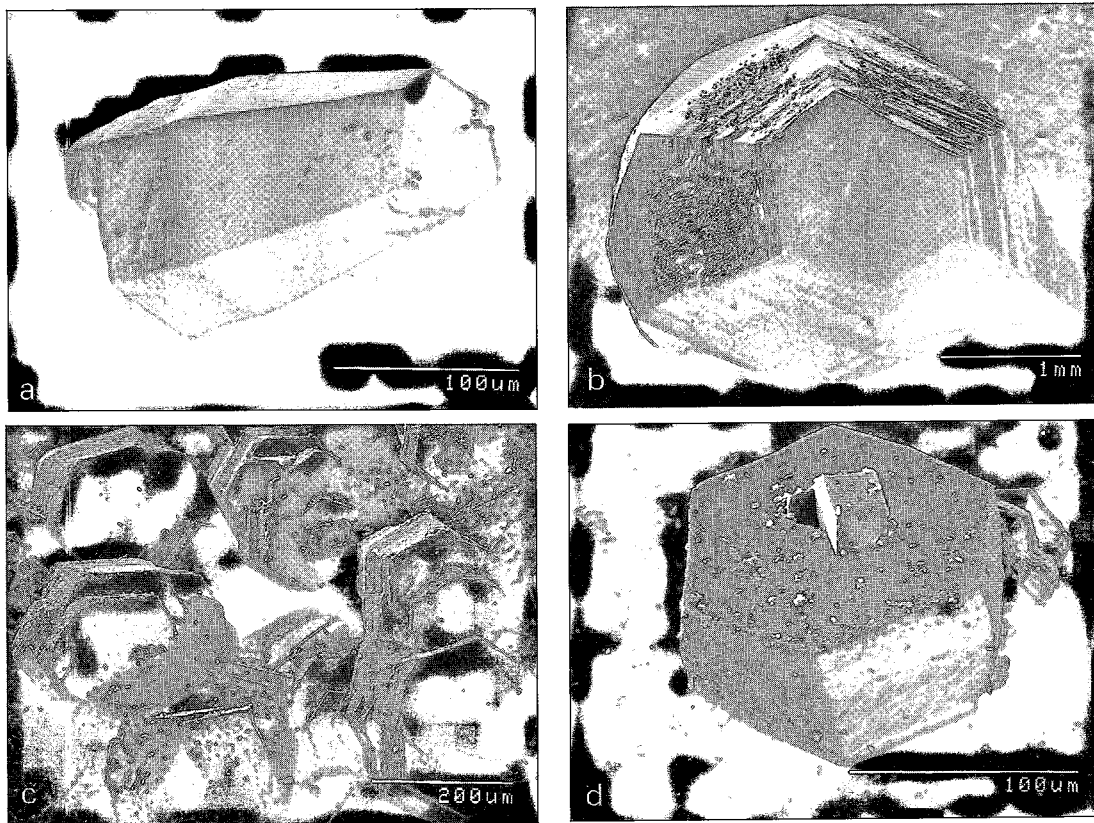


FIG. 1. Scanning electron micrographs of: a) charmarite-2H from Mont Saint-Hilaire, Quebec, b) quintinite-2H from the Jacupiranga mine, São Paulo, Brazil, c) caresite-3T from Mont Saint-Hilaire, Quebec, and d) caresite-3T from the Corporation quarry, Montreal, Quebec.

muscovite crystals. Caresite-3T was collected from the Poudrette quarry by Stephen and Janet Cares in 1989 and by one of the authors (GYC) in the late 1980s; it is associated with microcline, analcime, natrolite, calcite, a vermiculite- or smectite-group mineral, chamosite and aegirine. Caresite-3T also was found in 1955 by Dr. Peter Tarassoff in a construction excavation in the old Corporation quarry on Mont Royal, also one of the Monteregian Hills. This locality, situated in the city of Montreal, is now buried beneath a sports facility and is inaccessible. Here, caresite-3T occurs in small cavities in a nepheline syenite dike, in association with analcime, microcline, natrolite, tetranatrolite, aegirine, siderite, biotite, anatase, hematite, nordstrandite, dawsonite, berthierine, lovozerite, zircon, fluorite and pyrite. Quintinite-2H occurs in vugs in dolomitic carbonatite at the Jacupiranga mine, São Paulo, Brazil, and is associated with dolomite, magnetite, fluorapatite, phlogopite, pyrite, calcite, chalcopyrite and pyrrhotite. The material from this locality was originally described as manasseite (Menezes & Martins 1984).

#### PHYSICAL AND OPTICAL PROPERTIES

Charmarite-2H occurs as tabular hexagonal crystals to 0.5 mm across (Fig. 1a), commonly stacked along [0001] to form pagoda-like aggregates up to 1 mm long, and as thin mica-like plates in some cases forming star-shaped aggregates. The stacked tabular crystals can show parallel-growth features along [0001]. The most common forms observed are {0001} and  $\{10\bar{1}0\}$ . It is transparent, orange-brown, pale brown, pale blue or colorless. Charmarite-2H is optically negative, with weak pleochroism in pale-blue varieties,  $\omega$  = colorless,  $\epsilon$  = pale blue, and strong pleochroism in the orange-brown varieties,  $\omega$  = orange-brown,  $\epsilon$  = very pale brown. Charmarite-3T occurs as thin {0001} plates forming caps at the free end of prismatic crystals, or as stacked tabular crystals that taper off at the other end to give an overall appearance of a nail. The most common forms observed are {0001} and  $\{10\bar{1}0\}$ . It is transparent, orange-brown to pale brown, and pleochroic with  $\omega$  = brown,  $\epsilon$  = pale brown.

TABLE 1. SELECTED PROPERTIES OF CHARMARITE, QUINTINITE AND CARESITE

|                                  | CHARMARITE-<br>2H | CHARMARITE-<br>3T    | QUINTINITE-<br>2H | QUINTINITE-<br>3T    | CARESITE-<br>3T      |
|----------------------------------|-------------------|----------------------|-------------------|----------------------|----------------------|
| Space group                      | $P6_322$          | $P3_112$ or $P3_212$ | $P6_322$          | $P3_112$ or $P3_212$ | $P3_112$ or $P3_212$ |
| $a(\text{\AA})$                  | 10.985(5)         | 10.985(3)            | 10.571(1)         | 10.558(2)            | 10.805(3)            |
| $c$                              | 15.10(2)          | 22.63(3)             | 15.139(7)         | 22.71(3)             | 22.48(3)             |
| $V(\text{\AA}^3)$                | 1578(3)           | 2366(4)              | 1465(1)           | 2192(3)              | 2273(4)              |
| $Z$                              | 4                 | 6                    | 4                 | 6                    | 6                    |
| Optics                           | uniaxial (-)      | uniaxial (-)         | uniaxial (+)      | uniaxial(+/-)        | uniaxial (-)         |
| $\omega$                         | 1.587(1)          | 1.587(1)             | 1.533(1)          | 1.533(1)             | 1.599(1)             |
| $\epsilon$                       | 1.547(1)          | undetermined         | 1.533(1)          | 1.533(1)             | 1.570(1)             |
| $D_{\text{meas}} \text{ g/cm}^3$ | 2.47(1)           | 2.48(1)              | 2.14(1)           | 2.14(1)              | 2.59(1)              |
| $D_{\text{calc}} \text{ g/cm}^3$ | 2.50              | 2.50                 | 2.15              | 2.14                 | 2.59                 |

TABLE 2. SELECTED PROPERTIES OF HYDROTALCITE AND MANASSEITE

|                                  | HYDROTALCITE | MANASSEITE   |
|----------------------------------|--------------|--------------|
| Space group                      | $R\bar{3}m$  | $P6_3/mmc$   |
| $a(\text{\AA})$                  | 3.054        | 3.1          |
| $c$                              | 22.81        | 15.5         |
| $V(\text{\AA}^3)$                | 184.24       | 129.00       |
| $Z$                              | 3            | 1            |
| Optics                           | biaxial (-)  | uniaxial (-) |
| $\omega$                         | 1.511-1.531  | 1.524        |
| $\epsilon$                       | 1.495-1.529  | 1.510        |
| $D_{\text{meas}} \text{ g/cm}^3$ | 2.06         | 2.05         |
| $D_{\text{calc}} \text{ g/cm}^3$ | 2.16         | 2.00         |

Araokhova *et al.* (1996), Roberts *et al.* (1990).

Quintinite-2H occurs as equant crystals to 5 mm across and as prismatic crystals to 3 mm long (Fig. 1b) displaying the forms  $\{0001\}$  and an  $\{hkil\}$  trapezohedron with or without  $\{10\bar{1}0\}$ . Quintinite-2H crystals are transparent, ranging from a deep orange-red to orange to pale brown. Although the indices of refraction,  $\epsilon$  and  $\omega$ , are equal within the error of measurement ( $\pm 0.001$ ), elongation determinations show that the fast ray is always parallel to  $\omega$  and the slow ray is parallel to  $\epsilon$ , indicating the optic sign to be (+). It is pleochroic, with  $\omega =$  yellow,  $\epsilon =$  lighter yellow. Quintinite-3T occurs as hexagonal tabular crystals on  $\{0001\}$  to 1 mm across, commonly forming rosettes, in some cases in an epitactic relationship with donnayite-(Y). Common forms observed are  $\{0001\}$  and  $\{10\bar{1}0\}$ . Quintinite-3T is transparent, bright yellow to pale yellow. Like quintinite-2H, the indices of refraction,  $\epsilon$  and  $\omega$ , are equal within the error of measurement ( $\pm 0.001$ ), but elongation determinations show that both uniaxial (+) and (-) crystals are present on the same specimen. Most crystals observed are nonpleochroic; however, one with a Mg:Fe ratio of  $\sim 1$  is pleochroic, with  $\omega =$  dark green,  $\epsilon =$  light green.

Caresite-3T occurs as tabular crystals on  $\{0001\}$  (Figs. 1c, d) and pyramidal crystals to 0.5 mm, forming irregular aggregates. Forms observed are  $\{0001\}$  and  $\{10\bar{1}0\}$ , generally with a hexagonal pyramid, which is not consistent with the symmetry. This apparent form may be caused by a combination of an  $\{h0\bar{h}1\}$  trigonal dipyramid and an  $\{hh\bar{2}hl\}$  rhombohedron or by a diminishing pinacoid during crystal growth. Caresite-3T

is transparent and green-black, but is normally coated with a fine-grained smectite or with chamosite.

All members of this group have a white streak, vitreous luster and are nonfluorescent in ultraviolet light. All are brittle, with a Mohs hardness of about 2, have a perfect  $\{0001\}$  cleavage and an uneven fracture. Generally, the measured densities compare well with the calculated densities (Table 1). Twinning was observed only in the case of charmarite-3T, which is twinned by rotation of  $180^\circ$  about  $c$  to give 3H pseudosymmetry.

Selected properties of quintinite, charmarite and caresite are presented in Table 1, and those of hydrotalcite and manasseite are shown in Table 2 for comparison.

#### CHEMICAL COMPOSITION

Chemical analyses were done on (1) a Cambridge Microscan MK5 electron microprobe with an operating voltage of 15 kV and a beam current of 0.30  $\mu\text{A}$ , using the correction program EMPADR VII developed by Rucklidge & Gasparrini (1969), and modified by D. Marshall and P. Jones, and (2) a JEOL 733 Superprobe (wavelength-dispersion mode) with an operating voltage of 15 kV, a beam current of 0.20  $\mu\text{A}$ , and using Tracor-Northern automation with a conventional ZAF routine. Beam diameter varied from 10 to 50  $\mu\text{m}$  depending on the size of the grain. As broad a beam as possible was used to prevent sample burn-up. The following standards were used: synthetic  $\text{Fe}_2\text{O}_3$ , almandine and siderite (Fe), rhodochrosite (Mn), dolomite (Mg), anorthite and chlorite (Al) and LiF (F). The results of the electron-microprobe analyses, together with the number of atoms per formula unit (*apfu*) are listed in Table 3. The ideal formulae for the members of the group are given in Table 4.

Thermogravimetric (TG) and evolved-gas (EG) data were obtained from a modified Mettler TA-1 Thermoanalyzer and an attached Inficon IQ200 quadrupole mass spectrometer at the Royal Ontario Museum, Toronto (Wicks & Ramik 1990). Samples were lightly ground in HPLC-grade acetone and stored

TABLE 3. COMPOSITION OF CHARMARITE, QUINTINITE AND CARESITE

|                                | 1             |       | 2             |      | 3             |      | 4             |      | 5           |       | 6           |      |
|--------------------------------|---------------|-------|---------------|------|---------------|------|---------------|------|-------------|-------|-------------|------|
|                                | Charmarite-2H |       | Charmarite-3T |      | Quintinite-2H |      | Quintinite-3T |      | Caresite-3T |       | Caresite-3T |      |
|                                | wt%           | apfu  | wt%           | apfu | wt%           | apfu | wt%           | apfu | wt%         | apfu  | wt%         | apfu |
| MgO                            | 0.10          | 0.01  |               |      | 33.19         | 3.95 | 34.08         | 3.98 | 0.85        | 0.13  | 1.42        | 0.21 |
| FeO                            | 0.58          | 0.05  |               |      | 0.74          | 0.05 | 0.29          | 0.02 | 44.92       | 3.78  | 43.13       | 3.56 |
| Fe <sub>2</sub> O <sub>3</sub> |               |       | 0.26          | 0.02 | 1.15          | 0.07 | 0.80          | 0.05 |             |       |             |      |
| MnO                            | 47.07         | 3.99  | 46.53         | 4.01 |               |      |               |      | 1.00        | 0.09  | 1.71        | 0.14 |
| Al <sub>2</sub> O <sub>3</sub> | 17.21         | 2.03  | 16.49         | 1.98 | 20.52         | 1.93 | 21.14         | 1.95 | 17.05       | 2.02  | 17.68       | 2.06 |
| CO <sub>2</sub>                | 7.30          | 1.09  | 7.20          | 1    | 9.17          | 1    | 9.35          | 1    | 7.31        | 1     | 7.42        | 1    |
| H <sub>2</sub> O               | 26.70         | 17.43 | 26.54         | 18   | 33.80         | 18   | 34.43         | 18   | 26.84       | 17.95 | 28.33       | 18   |
| F                              | 0.10          | 0.03  |               |      |               |      |               |      | 0.17        | 0.05  |             |      |
| O=F                            | -0.04         |       |               |      |               |      |               |      | -0.07       |       |             |      |
| Total                          | 99.02         |       | 97.02         |      | 98.57         |      | 100.09        |      | 98.07       |       | 98.69       |      |

Charmarite-2H formula calculation based on 18 (O+F); all other formula calculations based on 7 oxygens, assuming stoichiometric CO<sub>2</sub>, H<sub>2</sub>O and OH.

1. Mont Saint-Hilaire, Québec. Average of 14 analyses. H<sub>2</sub>O and CO<sub>2</sub> determined by TGA-EGA.

2. Mont Saint-Hilaire, Québec. Average of 4 analyses.

3. Jacupiranga, São Paulo, Brazil. Average of 4 analyses. TGA-EGA shows a two-stage weight loss centered at 100°C (H<sub>2</sub>O) and 409°C (H<sub>2</sub>O and CO<sub>2</sub>) to give a total loss of 42.10 wt.%. However, CO<sub>2</sub> and H<sub>2</sub>O evolved at 409°C are not resolved and therefore were calculated by stoichiometry. The total calculated weight loss of CO<sub>2</sub> + H<sub>2</sub>O = 43.50 wt.%, close to the determined total weight loss of 42.10 wt.%. Total FeO = 1.77 and was partitioned to 2+ and 3+.

4. Mont Saint-Hilaire, Québec. Average of 3 analyses. Total FeO = 1.01 and was partitioned to 2+ and 3+.

5. Mont Saint-Hilaire, Québec. Average of 3 analyses.

6. Mont Royal, Montréal, Québec. Average of 4 analyses.

TABLE 4. IDEAL FORMULAS: CHARMARITE, QUINTINITE AND CARESITE

|                              |   |
|------------------------------|---|
| Charmarite-2H, charmarite-3T | Mn <sub>2</sub> Al <sub>2</sub> (OH) <sub>12</sub> CO <sub>3</sub> ·3H <sub>2</sub> O |
| Quintinite-2H, quintinite-3T | Mg <sub>2</sub> Al <sub>2</sub> (OH) <sub>12</sub> CO <sub>3</sub> ·3H <sub>2</sub> O |
| Caresite-3T                  | Fe <sub>2</sub> Al <sub>2</sub> (OH) <sub>12</sub> CO <sub>3</sub> ·3H <sub>2</sub> O |

at 79% RH. They were weighed directly in vacuum-baked Pt crucibles, subjected to high vacuum until weight-stable, and then heated at 10°C/minute. Owing to the scarcity of available material, the determination of H<sub>2</sub>O and CO<sub>2</sub> contents was attempted only for charmarite-2H from Mont Saint-Hilaire, Quebec, and quintinite-2H from São Paulo, Brazil. For charmarite-2H, TG-EG analysis showed a two-stage weight loss of H<sub>2</sub>O that was essentially complete at 376°C, giving a total loss of H<sub>2</sub>O of 26.7 wt.%. The first-stage weight loss, interpreted as loss of H<sub>2</sub>O, peaked at 81°C, and the second stage, interpreted as loss of OH, peaked at 262°C. The loss of CO<sub>2</sub> began at approximately 300°C, with smaller maxima at 400°, 538°, 635°C, and a more significant maximum at 739°C, and was essentially complete at 800°C. The total loss of CO<sub>2</sub> is 7.3 wt.%. For quintinite-2H, the TG curve shows strong maxima at 100° (loss of H<sub>2</sub>O), 409° (loss of OH and CO<sub>2</sub>) and weak maxima at 550°, 700° and 895°C, with a total weight loss of 42.1(1) wt.%. Unfortunately, there was a complete overlap of H<sub>2</sub>O and CO<sub>2</sub> evolution that rendered it impossible to estimate the weight loss of individual components. No gases other than CO<sub>2</sub> and H<sub>2</sub>O were detected by EG analysis, either in quintinite-2H or charmarite-3T.

There is extensive solid-solution between the Mg and Fe end-members. An intermediate member with Mg:Fe equal to 2.01:2.00 *apfu* has been found at

Mont Saint-Hilaire. Two analyses of a zoned crystal, also from Mont Saint-Hilaire, gave Mg:Fe equal to 2.64:1.33 and 3.30:0.66 *apfu*. However, Mn substitution for Fe in caresite-3T is rather restricted. The maximum Mn found in numerous analyses was 2.39 wt% MnO, or less than 0.2 *apfu*. Mn was not detected in the numerous analyses of quintinite-2H and quintinite-3T. The maximum Fe<sup>2+</sup> in the numerous analyses of charmarite-2H and charmarite-3T was found to be 1.05 wt.% FeO, or less than 0.1 *apfu* Fe<sup>2+</sup>.

The compatibility index  $1 - (K_p/K_c)$  for charmarite-2H, as derived from Gladstone-Dale calculations, is 0.038, which is classified as excellent (Mandirino 1981); for charmarite-3T,  $1 - (K_p/K_c)$  is 0.016 (superior), using only the value of  $\omega$ , as  $\epsilon$  was left undetermined because the crystals were too thin to measure in that direction; for quintinite-2H,  $1 - (K_p/K_c)$  is 0.008 (superior), for quintinite-3T, it is 0.011 (superior), and for caresite-3T, it is 0.030 (excellent).

Charmarite-2H and -3T are attacked by 1:1 HCl and HNO<sub>3</sub> with effervescence, and dissolve in 1:1 H<sub>2</sub>SO<sub>4</sub> with effervescence. Quintinite-2H is attacked slightly by 1:1 HCl, more strongly by 1:1 HNO<sub>3</sub>, and very strongly by 1:1 H<sub>2</sub>SO<sub>4</sub>, all with effervescence. Quintinite-3T is attacked slightly by 1:1 HCl, strongly by HNO<sub>3</sub>, and dissolves in 1:1 H<sub>2</sub>SO<sub>4</sub>. Caresite-3T dissolves in 1:1 HCl, and is strongly attacked by 1:1 HNO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub>, with effervescence.

#### INFRARED ANALYSIS

The infrared spectra (Fig. 2) of all members of the group were obtained using a Bomem Michelson MB-120 Fourier-transform infrared spectrometer with a diamond-anvil-cell microsampling device. The spectra are very similar, with a broad double peak in the 3100 - 3400 cm<sup>-1</sup> region, which represents the

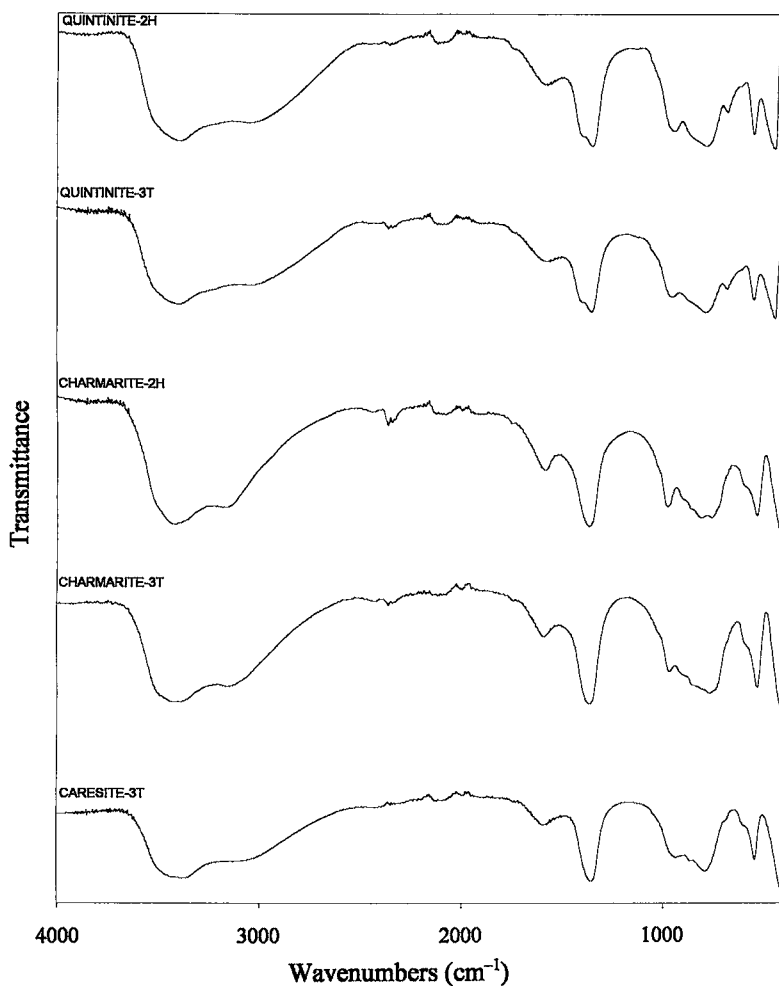


FIG. 2. Infrared spectra of the new species quintinite-2H, quintinite-3T, charmarite-2H, charmarite-3T and caresite-3T.

O–H stretching frequencies of H<sub>2</sub>O and OH groups. H<sub>2</sub>O is confirmed by the presence of the H–O–H bending mode at 1580 cm<sup>-1</sup>. The peak at 1360 cm<sup>-1</sup> is assigned to the asymmetric stretching mode of CO<sub>3</sub> ( $\nu_3$ ). The peaks at 400–950 cm<sup>-1</sup> are complicated, and could not be unequivocally assigned, although the peak in the region of 950 cm<sup>-1</sup> may be assigned to the OH-bending mode. The  $\nu_2$  and  $\nu_4$  modes of CO<sub>3</sub> likely contribute to the broad peak centered at 785 cm<sup>-1</sup>. The presence of a shoulder at 1400 cm<sup>-1</sup> on the quintinite-2H and -3T spectra may indicate a lowering in the ideal C<sub>3v</sub> symmetry of the carbonate group (Hernandez-Moreno *et al.* 1985). Furthermore, they suggested that the lowering of symmetry of the carbonate group would result in the

activation of the  $\nu_1$  (symmetric stretching) mode of CO<sub>3</sub>. The small peak in the region of 1135 cm<sup>-1</sup> in the quintinite-2H and -3T spectra seems to confirm this. The main difference between the spectra of the 4:2 minerals (this study) and the 6:2 minerals (Ferraro 1982, Jones & Jackson 1993) seems to be the lowering of the frequencies in the OH stretching and H–O–H bending modes, and of the broad peak in the 600–800 cm<sup>-1</sup> region.

#### X-RAY CRYSTALLOGRAPHY

Single-crystal X-ray precession studies show the 2H form of the 4:2 minerals to be hexagonal, space group

TABLE 5. X-RAY POWDER-DIFFRACTION DATA:  
QUINTINITE, CHARMARITE AND CARESITE

| QUINTINITE-3T |                              |                               |            | QUINTINITE-2H |                              |                               |            |
|---------------|------------------------------|-------------------------------|------------|---------------|------------------------------|-------------------------------|------------|
| <i>I</i>      | $d_{\text{obs}}(\text{\AA})$ | $d_{\text{calc}}(\text{\AA})$ | <i>hkl</i> | <i>I</i>      | $d_{\text{obs}}(\text{\AA})$ | $d_{\text{calc}}(\text{\AA})$ | <i>hkl</i> |
| 100           | 7.57                         | 7.570                         | 003        | 100           | 7.63                         | 7.569                         | 002        |
| 90            | 3.778                        | 3.7850                        | 006        | 100           | 3.785                        | 3.7847                        | 004        |
| 5             | 2.622                        | 2.6218                        | 221        | 15            | 2.603                        | 2.6035                        | 221        |
| 40            | 2.570                        | 2.5709                        | 222        | 15            | 2.496                        | 2.4952                        | 222        |
| 5             | 2.531                        | 2.5308                        | 305        | 15            | 2.341                        | 2.3412                        | 223        |
| 40            | 2.281                        | 2.2821                        | 225        | 15            | 2.166                        | 2.1668                        | 224        |
| 40            | 1.932                        | 1.9330                        | 228        | 15            | 1.991                        | 1.9910                        | 225        |
| 10            | 1.721                        | 1.7215                        | 22.10      | 20            | 1.825                        | 1.8250                        | 226        |
| 5             | 1.627                        | 1.6262                        | 22.11      | 10            | 1.537                        | 1.5386                        | 228        |
| 20            | 1.524                        | 1.5239                        | 600        | 10            | 1.526                        | 1.5286                        | 600        |
| 20            | 1.493                        | 1.4939                        | 603        | 15            | 1.495                        | 1.4958                        | 602        |
| 5             | 1.457                        | 1.4568                        | 22.13      | 5             | 1.416                        | 1.4151                        | 604        |
| 10            | 1.415                        | 1.4136                        | 606        | 10            | 1.3140                       | 1.3136                        | 22.10      |
|               |                              |                               |            | 5             | 1.3018                       | 1.3017                        | 442        |
|               |                              |                               |            | 5             | 1.2789                       | 1.2783                        | 443        |
|               |                              |                               |            | 5             | 1.2619                       | 1.2616                        | 00.12      |
|               |                              |                               |            | 5             | 1.2204                       | 1.2207                        | 22.11      |
|               |                              |                               |            | 10            | 1.1705                       | 1.1707                        | 446        |
| CHARMARITE-3T |                              |                               |            | CHARMARITE-2H |                              |                               |            |
| <i>I</i>      | $d_{\text{obs}}(\text{\AA})$ | $d_{\text{calc}}(\text{\AA})$ | <i>hkl</i> | <i>I</i>      | $d_{\text{obs}}(\text{\AA})$ | $d_{\text{calc}}(\text{\AA})$ | <i>hkl</i> |
| 100           | 7.55                         | 7.545                         | 003        | 100           | 7.53                         | 7.551                         | 002        |
| 90            | 3.770                        | 3.7727                        | 006        | 60            | 3.768                        | 3.7755                        | 004        |
| 70            | 2.670                        | 2.6690                        | 222        | 10            | 2.743                        | 2.7464                        | 220        |
| 5             | 2.469                        | 2.4709                        | 224        | 10            | 2.702                        | 2.7021                        | 221        |
| 70            | 2.346                        | 2.3482                        | 225        | 50            | 2.578                        | 2.5810                        | 222        |
| 5             | 2.092                        | 2.0933                        | 227        | 10            | 2.410                        | 2.4109                        | 223        |
| 60            | 1.973                        | 1.9708                        | 228        | 40            | 2.221                        | 2.2209                        | 224        |
| 10            | 1.887                        | 1.8864                        | 00.12      | 10            | 2.031                        | 2.0320                        | 225        |
| 15            | 1.747                        | 1.7468                        | 22.10      | 40            | 1.856                        | 1.8556                        | 226        |
| 10            | 1.645                        | 1.6469                        | 22.11      | 20            | 1.585                        | 1.5856                        | 600        |
| 30            | 1.586                        | 1.5857                        | 600        | 40            | 1.552                        | 1.5517                        | 602        |
| 30            | 1.662                        | 1.5518                        | 603        | 5             | 1.462                        | 1.4619                        | 604        |
| 5             | 1.464                        | 1.4618                        | 606        | 5             | 1.3252                       | 1.3233                        | 22.10      |
| 10            | 1.2577                       | 1.2577                        | 22.16      | 5             | 1.2908                       | 1.2905                        | 444        |
|               |                              | 1.2576                        | 00.18      |               |                              |                               |            |
| CARESITE-3T   |                              |                               |            |               |                              |                               |            |
| <i>I</i>      | $d_{\text{obs}}(\text{\AA})$ | $d_{\text{calc}}(\text{\AA})$ | <i>hkl</i> |               |                              |                               |            |
| 100           | 7.49                         | 7.494                         | 003        |               |                              |                               |            |
| 50            | 3.746                        | 3.7449                        | 006        |               |                              |                               |            |
| 40            | 2.625                        | 2.6265                        | 222        |               |                              |                               |            |
| 50            | 2.314                        | 2.3156                        | 225        |               |                              |                               |            |
| 40            | 1.948                        | 1.9475                        | 228        |               |                              |                               |            |
| 10            | 1.729                        | 1.7281                        | 22.10      |               |                              |                               |            |
| 10            | 1.631                        | 1.6299                        | 22.11      |               |                              |                               |            |
| 15            | 1.558                        | 1.5596                        | 600        |               |                              |                               |            |
| 20            | 1.526                        | 1.5269                        | 603        |               |                              |                               |            |
| 5             | 1.458                        | 1.4565                        | 22.13      |               |                              |                               |            |
| 5             | 1.440                        | 1.4398                        | 606        |               |                              |                               |            |
| 5             | 1.3818                       | 1.3804                        | 22.14      |               |                              |                               |            |
| 5             | 1.2478                       | 1.2491                        | 00.18      |               |                              |                               |            |

Gandolfi 114.6 mm diameter camera, CuK $\alpha$  radiation,  $\lambda = 1.5418$ , visual intensities.

$P6_322$ , and the 3T form to be trigonal, space group  $P3_112$  or  $P3_212$ . The cell parameters (Table 1) were derived from single-crystal photographs and refined by a least-squares method using X-ray powder-diffraction data (Table 5). The powder data for hydrotalcite and manasseite are presented in Table 6 for comparison. Unlike the 6:2 minerals, the single-crystal photographs of both 2H and 3T forms of the 4:2 minerals show well-defined superlattice reflections that give rise to a strong subcell with  $a' = \frac{1}{3} a \sin 60^\circ \approx 3.05 \text{\AA}$  and  $c' = c$ .

The subcells of these 4:2 minerals resemble the cells of the corresponding 6:2 minerals ( $a = 3.1$ ,  $c = 15.6$  or  $23.4 \text{\AA}$ ; Taylor 1973) and have the same space-group symmetry,  $P6_3/mmc$  and  $R\bar{3}m$ , indicating a close structural relationship between the two groups of minerals. The hexagonal trapezohedron observed as a prominent form on crystals of quintinite-2H from São Paulo, Brazil, indicates unequivocally that the crystals are 622, which is isomorphous with the space group derived for the 2H forms of the 4:2 minerals.

TABLE 6. X-RAY POWDER-DIFFRACTION DATA: HYDROTALCITE AND MANASSEITE

| HYDROTALCITE |              |            | MANASSEITE |              |            |
|--------------|--------------|------------|------------|--------------|------------|
| <i>l</i>     | <i>d</i> (Å) | <i>hkl</i> | <i>l</i>   | <i>d</i> (Å) | <i>hkl</i> |
| 100          | 7.69         | 006        | 100        | 7.67         | 002        |
| 70           | 3.88         | 018,0,0.12 | 20         | 3.83         | 004        |
| 20           | 2.58         | 024        | 10         | 3.71         | 103        |
| 20           | 2.30         | 0210       | 50         | 2.60         | 201,113    |
| 20           | 1.96         | 125,0216   | 30         | 2.49         | 202        |
| 10           | 1.85         | 2110       | 40         | 2.34         | 203        |
| 10           | 1.75         | 2113,3020  | 40         | 2.17         | 204,115    |
| 10           | 1.65         | 2116       | 40         | 2.00         | 210,205    |
| 20           | 1.53         | 220,223    | 60         | 1.84         | 206        |
| 20           | 1.50         | 226        | 20         | 1.56         | 208        |
| 10           | 1.28         | 4010       | 30         | 1.52         | 221        |
|              |              |            | 30         | 1.49         | 119        |
|              |              |            | 10         | 1.42         |            |
|              |              |            | 10         | 1.33         |            |
|              |              |            | 10         | 1.25         |            |
|              |              |            | 10         | 1.24         |            |
|              |              |            | 10         | 1.17         |            |
|              |              |            | 10         | 1.09         |            |

Ross &amp; Kodama (1967).

## DISCUSSION

The Mn and Fe end-members of the 4:2 minerals are unique, whereas the Mg end-member is not entirely new. Hydrotalcite was originally established as a mineral species by Manasse in 1915 (Frondel 1941), with the composition  $Mg_6Al_2(OH)_{16}CO_3 \cdot 4H_2O$ ; it has since been reported from a number of localities (Frondel 1941, Roberts *et al.* 1990). Černý (1963) reported a rhombohedral mineral from a quartz-oligoclase pegmatite in a serpentinite at Věžná, western Moravia, Czech Republic, with Mg:Al close to 2:1, and called it hydrotalcite. A crystal-structure determination of hydrotalcite was done on a 4:2 crystal from this locality (Allmann & Jepsen 1969). A hexagonal 4:2 mineral with a composition identical to that of quintinite-2H was found in the evaporite deposits of the pre-Caspian depression and in those of central Asia (Drits *et al.* 1986, 1987), and the name *manasseite* was given to it. As the cell contents for the Mg-Al 6:2 minerals are less than one formula unit ( $Z = 0.25$  for the 2H form, and  $Z = 0.375$  for the 3R form), and the Mg:Al ratio may vary in synthetic hydrotalcite and hydrotalcite-like materials, the general formula  $[Mg_{1-x}Al_x(OH)_2]^{2+}[A_{x/n}^{n-} \cdot mH_2O]$ , or some variant of it, is frequently used to represent these compounds (Brindley & Kikkawa 1980, Mascolo & Marino 1980, Serna *et al.* 1982, Miyata 1983, Drits *et al.* 1986, Pausch *et al.* 1986). In this general formula, A is an exchangeable anion with valence of  $n$ , and  $x$  is in the range 0.20–0.33 according to most authors. However, for minerals,  $x$  seems to be fixed at 0.25 in the 6:2 minerals and 0.33 in the 4:2 minerals. Intermediate compositions have not been reported. The  $<1$  value of  $Z$  in the 6:2 minerals represents the smallest subcell in the disordered structure. The true cell in the ordered 4:2 minerals is several times larger.

Allmann & Lohse (1966), Ingram & Taylor (1967), Allmann & Jepsen (1969) and Taylor (1973) showed that the structure of the 6:2 minerals is based on positively charged brucite-like layers with interlayered carbonate groups and  $H_2O$  molecules. In the brucite-like layers, the octahedrally coordinated divalent and

trivalent cations are fully disordered, whereas in the interlayers, the O atoms of carbonate groups and  $H_2O$  molecules are statistically distributed over a single set of sites. In the numerous studies of the 6:2 minerals, superlattice reflections have not been observed. In contrast, well-defined superlattice reflections occur in the 4:2 minerals and strongly suggest the presence of cation ordering. The fact that the superlattice reflections are much weaker for the Mg minerals than for the Mn and Fe minerals further suggests that this ordering occurs at the octahedral sites of the brucite-like layers. This interpretation is consistent with results of infrared studies of synthetic hydrotalcite-like compounds (Hernandez-Moreno *et al.* 1985), which show that ordering of octahedrally coordinated cations is present only in the 4:2 compounds and not in the 6:2 compounds. In their recent paper on the crystal chemistry of  $Al_2Mg_4(OH)_{12}(CO_3) \cdot 3H_2O$ , Arakcheeva *et al.* (1996) stated that a crystal-structure analysis on the material from São Paulo, Brazil (quintinite-2H) shows complete order of all atoms.

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